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(54) **Polymeric compositions and films, plural layer sheet structures and packaging made from such films and structures.**

(57) A polymeric composition is a blend of a first component comprising polypropylene, a second component comprising an ethylene-based copolymer, and a third component which may be an elastomeric polymer. The blends are useful in making single and plural layer sheet structures. Such a structure comprises a first layer (12) made from said blend and a second layer (14) of an abuse-resistant polymer such as polypropylene. Optionally, the structure includes further layers, e.g. a barrier layer to prevent or reduce gas infusion. The blend layer (12) can be employed as the means for making heat seals (28) between layers of the sheet structure to form a packaging pouch (30), and is able to withstand retorting conditions without becoming significantly embrittled.

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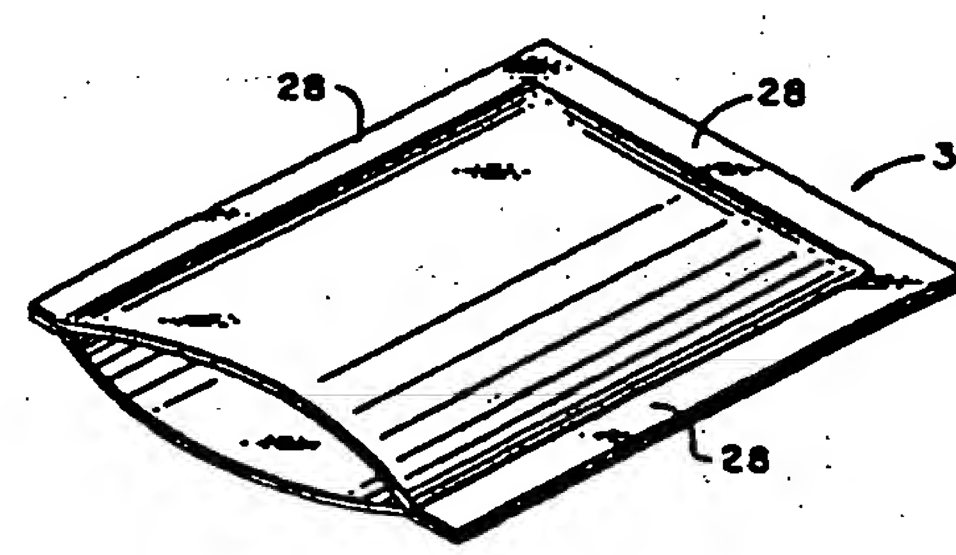
FIG.1

FIG.5



POLYMERIC COMPOSITIONS AND FILMS, PLURAL LAYER SHEET STRUCTURES AND PACKAGING MADE FROM SUCH FILMS AND STRUCTURES

The present invention relates to polymeric compositions and films, plural layer sheet structures and packaging made from such films and structures.

The packaging industry makes and uses many sheet stuctures in the making of packages such as heat sealed packages. Such packages find use in a multiplicity of applications. Of particular interest herein are those applications where the finished package may be subjected to relatively abusive physical shocks, for example by being dropped, bumped, or the like. In cases where the package contains fluid components, the shock may be transferred to all parts of the package by the contained fluid. The transferred shock can be effective to cause failure of the package at its weakest point, when the severity of shock is sufficiently great, even though the initial shock is not exerted at the weakest point.

Heat sealed packages tend to fail at or close to a heat seal. Typical failures occur in the seal area by partial peeling of the facing sealant layers from each other. Where the shock is severe enough, the package may have a catastophic failure wherein the package may be penetrated to the outside.

Such failure of the package, whether by partial peeling or by catastrophic failure, compromises the containment and protective functions of the package, and is thus not acceptable. It is desirable to find material compositions which may be used in packaging sheet structures to enhance the capability of the sheet structure, and particularly the sealant layer, to withstand increased amounts of shock-type abuse.

In certain packaging applications, the packages are subjected to heat treatment at elevated temperatures of up to about 250 °F (121 °C) as part of the packaging process. Such heat treatments are common in certain packaging of medical supplies and shelf stable foods.

Some polymers, such as polypropylene, which are otherwise excellent for use in high temperature applications, tend to be embrittled after being subjected to such heat treatments. Increasing brittleness leads to a reduced ability to resist shock type abuse. While the use of such materials is desirable for their ability to withstand heat treatment without failure of the package during the heat treatment, the resulting brittleness is an undesirable feature as it affects its ability to withstand the shocks of, for example, shipping and handling.

It is an object of this invention to provide novel material blend compositions, single layer films and plural layer films and sheet structures, incorporating the novel compositions, for use in making packages, and particularly heat sealed packages.

It is desired, also, to provide heat sealed packages capable of withstanding increased amounts of physical shock type abuse, especially after the closed and sealed package has been subjected to elevated temperatures of up to about 250 °F (121 °C).

According to the present invention, there is provided a multiple layer sheet structure comprising:

(a) a first layer, the composition of said first layer having (i) a first component comprising a polypropylene polymer, (ii) as a second component, a polymeric composition providing elastomeric properties, said second component comprising a first moiety of ethylene and a second moiety chosen from the group consisting of propylene and butene-1, and (iii) a third component, the composition of said third component being a modifier providing elastomeric properties; and

(b) a second layer adhered to said first layer, the composition of said second layer being selected from the group consisting of polymers, paper, and metal foil.

Further, there is provided a composition of matter which is a polymeric blend, comprising:

(a) 40 to 70% eg. 60% to 70% by weight of a first component, comprising a polypropylene polymer;

(b) a second component, comprising an ethylene-based copolymer of ethylene and a second moiety comprising either propylene or butene-1; and

(c) a third component, different from the second component which provides elastomeric properties.

In the above-defined sheet structure and blend composition, the second component is an ethylene-based copolymer (hereinafter sometimes referred to as "EBC") which consists essentially of ethylene copolymerized with moieties of a second molecule. The composition of the second molecule is selected from the group consisting of propylene and butene-1. Preferably the EBC consists essentially of an ethylene copolymerized with moieties of a second molecule, the copolymer being about 10 to about 95 mole percent ethylene and about 90 to about 5 mole percent of the moieties of the second molecule, the second molecule as stated being selected from propylene and butene-1. The aforesaid third component is an EBC or an elastomeric polymer. (Hereinafter, the aforesaid third component group of materials is sometimes referred to as

the "third component").

Preferably, the second component is present in an amount of about 5% to about 35% by weight of the overall composition of the blend oF which it forms a part.

In one preferred embodiment, the ethylene-based copolymer is essentially a copolymer of about 85 to about 95 mole percent ethylene and about to about 5 mole percent butene-1. In another embodiment, it is a copolymer of 81 - 94 mole percent ethylene and 19 - 6 mole percent of butene-1.

In another preferred embodiment, the ethylene-based copolymer is essentially a copolymer of about 60 to about 80 mole percent ethylene and about 40 to about 20 mole percent propylene.

The first component is preferably present in an amount of about 40% to about 70% by weight of the overall composition of the blend, and the third component is preferably present in an amount of about 10% to about 40% by weight. It is preferred that the composition of the third component be selected from the group of elastomers comprising polybutylene, polyisobutylene, ethylene propylene diene monomer terpolymer, styrene butadiene styrene copolymer, styrene ethylene butylene styrene copolymer, polybutene- 1, styrene isoprene styrene copolymer, isobutylene isoprene copolymer, or from the ethylene-based copolymers group.

In one family of preferred compositions, therefore, the first component is present in an amount of about 40% to about 70% by weight, the second component is present in an amount of about 5% to about 35% by weight, and the third component is present in an amount of about 10% to about 40% by weight. In that family of overall compositions, the composition of the third component is most desirably selected from the styrene butadiene styrene copolymer, styrene ethylene butylene styrene copolymer, styrene isoprene styrene copolymer, and polyisobutylene.

The aforesaid composition forms the basis of single layer films, plural layer films and multiple layer sheet structures embodying the invention.

In one multiple layer sheet structure according to the invention, a first layer is made of metal foil having two opposing surfaces on opposing sides thereof, a second layer of an abuse resistant polymer is adhered to one of the foil surfaces, and a third layer adhered to the sheet structure on the other of the foil surfaces, the composition of the third layer being a novel blend composition according to the invention as defined hereinbefore.

In another multiple layer sheet structure according to the invention, a first layer is made of a polyamide having two opposing surfaces on opposing sides thereof, a second layer of an ethylene vinyl alcohol copolymer having two opposing surfaces is disposed with one surface of the second layer on one surface of the first layer, a third layer of a polyamide is disposed on the other surface of the second layer opposite the first layer, and a fourth layer is adhered to the sheet structure on the other side of the first layer, the fourth layer being a novel blend composition according to the invention.

In yet another multiple layer sheet structure of the invention, a first layer is made of a barrier material having two opposing surfces on opposing sides thereof, the composition of the first layer being a vinylidene chloride copolymer, a second layer made of an abuse resistant polyner is adhered to one surface of the first layer, and a third layer is adhered, in the sheet structure on the other side of the first layer, the composition of the third layer being a novel blend according to the invention.

Packages of various types such as pouches or sachets, which may or may not be heat sealed, may also be made of the films and sheet structures according to the invention. The invention comprehends these packages when ready for filling and after filling and closing.

Embodiments of the invention will now be explained in more detail in the following description, which is given by way of example only and with reference to the accompanying drawings, in which:

FIGURE 1 is a cross-section of a portion of a 2-layer film according to the invention,

FIGURE 2 is a cross-section of a portion of a 3-layer sheet structure according to the invention, wherein a layer of a metal foil is incorporated as a barrier layer,

FIGURE 3 is a cross-section of a portion of a 4-layer film according to the invention,

FIGURE 4 is a cross-section of a portion of another 3-layer film according to the invention, wherein a polymeric barrier material is incorporated as an internal layer, and

FIGURE 5 is a perspective view of a typical pouch made in accordance with the invention.

The invention encompasses a plurality of forms and embodiments. In its most generic form, the invention lies in a composition of matter formed by the intimate blending together of several blend components. The invention may also be represented by a variety of articles which can be formed such as by molding or extruding the blend composition. One preferred form of article so fabricated is a single layer packaging film which may be formed by conventional extrusion processes. Such films are typically thin and flexible, being of the order of about 1 mil to about 8 mils (0.025 to 0.20 mm) thick. Thicker sections and shapes may also be formed, as for other uses.

Other preferred embodiments of the invention are those where the blend composition is used to

form one layer of a plural or multiple layer sheet structure. Representative of these films are those illustrated in FIGURES 1-4. Other sheet structures are possible, and contemplated. Sheet structures according to the invention have two, three, four or more than four, e.g. five, separate layers adhered one to another, using further intervening layers of adhesive as necessary.

Turning now to FIGURE 1, layer 12 is a layer of a novel blend composition according to the invention and layer 14 is formed of an abuse resistant polymer, such as polypropylene. A single layer film according to the invention will have a composition equivalent to the said layer 12.

The structure of FIGURE 1 illustrates a simpler form of packages of the invention. It is advantageous in its simplicity, and may find use in applications where high levels of protection from gaseous infusion are of paramount importance.

Where protection from gaseous infusion is more important, a layer of a barrier material is used. In the three layer structure of FIGURE 2, the additional layer 16 is a layer of metal foil, which provides an excellent barrier to gaseous infusion. Layer 14 is the layer of an abuse resistant polymer and layer 12 is the blend composition layer as in FIGURE 1.

The structure of FIGURE 2 is representative of a sheet structure useful in making retortable pouches. Such structures may also be used for applications where the package is not subjected to retort conditions. In those cases, the specific materials selected for each layer need not be evaluated in terms of their ability to withstand the retorting process. Typical problems encountered in retort processing are excessive softening during the processing, or embrittlement resulting from the processing. The foil layer 16 of such a retortable package is commonly 28 gauge to 100 gauge, with the thinner gauges being preferred for economic reasons. The abuse resistant layer 14 may be any of the conventionally known abuse resistant polymers. These include, for example, nylon, oriented nylon, oriented poly-propylene, and oriented polyester. For use in retort packaging, layer 14 is typically about 50 gauge, with a range of about 25 gauge to about 100 gauge. It is conventionally known to achieve adhesion between the foil layer 16 and abuse resistant layer 14 by means of adhesives such as polyester urethane curing type adhesives. These adhesives are acceptable for such use herein.

The blend layer 12 may be any of the blend compositions according to the invention. The FIGURE 2 structure is designed such that layer 12 may be used as a heat seal layer. A minimum thickness for achieving strong heat seals is about 1 mil (0.025 mm), so that is a preferred, minimum thickness. Thicker heat seal layers may advantageously provide more efficient use in heat seal material, up to about 4 mils (0. 10 mm); and that is a generally preferred maximum thickness. While layer 12 may be much thicker, for example about 8 mils (0.20 mm) or more, and same is encompassed by the scope of the invention, no material benefit is usually derived from the use of the additional material; so the greater thicknesses are not generally preferred.

Layer 12 may be adhered to layer 16 by a conventional polyester urethane curing type adhesive Alternately, adhesion may be achieved by other methods such as by extrusion lamination, coextrusion lamination, or extrusion coating followed by heat and pressure at a hot nip. In some instances, it may be desirable to use a primer on the surface of foil layer 16 before adhering layers 12 and 16 to each other.

Turning next to FIGURE 3, layer 12 is a blend composition according to the invention as in FIGURES 1 and 2. Layer 18 is a polyamide. Layer 20 is an ethylene vinyl alcohol copolymer. Layer 22 is a polyamide.

The structure of FIGURE 3 represents generically a family of sheet structures which provide an effective barrier to transmission of oxygen through the sheet structure while affording tansparency of the packaging material for visibility of the packaged contents. These packages have a variety of uses, and the selection of specific polymers and combinations of polymers depends on the particular use contemplated. A typical use is, as for the structure of FIGURE 2, that of retort packaging.

It is known to coextrude a multiple layer film structure wherein a layer of ethylene vinyl alcohol copolymer is between two layers of polyamide. Such a structure is seen in the substructure of layers 18, 20 and 22. It has further been found acceptable to include layer 12 as part of the coextrusion. Thus, the structure of FIGURE 3 may be advantageously and economically produced in a single processing step by coextruding all four layers simultaneously and combining them in the coextrusion process to form the multiple layer sheet structure.

For enhanced adhesion between layers 12 and 18, it is sometimes desirable to include an adhesive, such as an adhesive polymer, between layers 12 and 18. Such materials are conventionally known and may be selected based on the specific compositions of layers 12 and 18 and the process to be used. Typical of these adhesive polymers are those based on polypropylene and having carboxyl modifications thereto, such as maleic acid or maleic anhydride.

The structure may, of course, be made by other processes but they generally will be more

expensive, and thus those processes are less preferred. Illustrative of these is adhesive lamination wherein the layers may be joined by using, for example, polyester urethane curing type adhesive.

The term polyamide, as used in describing the compositions of layers 18 and 22 is intended to include copolymers and alloys of polyamide as a major component. Likewise, additives which are normally used with polyamide are acceptable herein and are thus included in the compositions of layers 18 and 22. While the compositions of layers 18 and 22 are usually the same, to facilitate coextrusion of the sheet material, they may be different and the process may be adapted accordingly.

The term ethylene vinyl alcohol copolymer as used in describing the composition of layer 20, is intended to include the copolymer alone and blends of ethylene vinyl alcohol with other polymers. Likewise, additives which are normally used with ethylene vinyl alcohol copolymer are acceptable herein and are thus included in the composition of layer 20.

The term "elastomeric" includes polymers, copolymers, rubbers, and the like which at room temperature can be stretched substantially under low stress and, upon immediate release of the stress, have the tendency to return generally to approximately the original shape.

The blend layer 12 may be any of the blend compositions embodying the invention. The FIGURE 3 structure, as in FIGURES 1 and 2, is designed such that layer 12 may be used as a heat seal layer. A minimum thickness for achieving strong heat seals is likewise about 1 mil (0.025 mm) in this structure; so that is a preferred minimum thickness. Thicker heat seal layers may advantageously provide more efficient use of the heat seal material, up to about 4 mils (0.10 mm); so that is a generally preferred maximum thickness. Layer 12 may also be thicker, as described for the corresponding layer 12 in FIGURE 2.

Layer 12 may also be less than 1 mil thick (0.025 mm) as, for example, in all the illustrated embodiments, where heat seals are not used or seal strength is not critical.

Turning now to FIGURE 4, layer 14 is an abuse resistant layer as in FIGURE 2. Layer 12 is of a blend composition according to the invention as in FIGURES 1-3. Layer 24 is an oxygen barrier layer such as a vinylidene chloride copolymer. Preferred copolymers are vinylidene chloride-vinyl chloride copolymer and vinylidene chloride-methyl-acrylate copolymer. In the FIGURE 4 structure, layer 24 usually has a thickness of 0.3 to 2.0 mil (0.008 to 0.051 mm), depending primarily on the rate of oxygen transmission which is desired for any given end use. While adhesion may be achieved by other means, suitable adhesives, such as the polyester urethane curing adhesives, are usually economically used between the respective layers 12, 24, and 14 to achieve good adhesion between the layers.

For constructing the sheet structure of FIGURE 4, layers 12, 24 and 14 are usually formed separately from each other and then combined into the multiple layer structure shown, by conventional techniques for combining polymer films. In a typical process, the layers are joined by conventional adhesive lamination techniques.

As is now evident from the plurality of the multiple layer structures illustrated above, the blend composition of polypropylene and the second and third components has useful advantage in a variety of multiple layer structures. Indeed a layer of the blend composition may advantageously be used in combination with any other layer which may be adhered to it. Thus the multiple layer embodiment of the invention may be generically defined as a multiple layer sheet material wherein the composition of at least one layer is comprised of a blend of polypropylene and the second and third components; and wherein the composition of the other layer or layers need not necessarily be restricted except for purposes of obtaining interlayer adhesion acceptable to the intended use. Thus non-polymeric materials, such as paper and metal foil, and the like, may be used, as well as polymeric materials.

The multiple layer sheet structures of the invention are advantageously formed into packages using heat seals, as illustrated in FIGURE 5. Typically, portions of the sheet material are brought into face-to-face relationship, with the layer 12 on one portion facing the layer 12 of the opposing sheet portion. Heat seals 28 are then formed between the sheet portions about a common periphery to form a container 30 that generally defines an enclosed area. Usually, one side of the thus formed package is left open, as shown, for insertion of the product. Finally, the filled package is closed and sealed by a heat seal along the open side.

Alternately, the sheet may be formed into a tube by forming a longitudinal seal along overlapping edges; and a transverse seal is formed across the width of the tube; all as described, for example, in US-A-4,521,437, herein incorporated by reference. This process is particularly suitable for use in vertical form, fill and seal machines.

In overcoming the primary problem of brittleness of the polypropoylene in flexible packages, as addressed herein, a number of factors work together in combination. Important to achievement of the objectives of the invention is incorporation, into the blend composition, of the second component and third components as hereinbefore described. The combined contribution of the second and third components is evidenced by substantially

fewer structural failures in the films, sheet structures, and packages made therefrom.

The ethylene-based copolymer of the second component is essentially a copolymer of two moieties, both moieties being present in the main chain of the polymer. The overall mole ratio is about 10 to about 95 mole percent ethylene, and conversely about 90 to about 5 mole percent of the second moiety. The ethylene is preferably copolymerized with propylene or butene-1. In the case of propylene, the mole ratio is most preferably within the range of about 20% to about 40% propylene and about 80% to about 60% ethylene. One such material is sold by Mitsui Petrochemical Industries, Ltd. as TAFMER P. In the case of butene-1, the mole ratio is most preferably within the range of about 85% to about 95% ethylene, and about 15% to about 5% butene-1. One such material is sold by Mitsui Petrochemical Industries, Ltd. as TAFMER A.

If an elastomeric polymer is used as the third component, the same may be any of the known and readily available elastomeric polymers, indeed even blends thereof. Examples of readily available elastomeric polymers are polybutylene, polyisobutylene, ethylene propylene diene monomer terpolymer, styrene butadiene styrene copolymer, styrene ethylene butylene styrene copolymer, styrene isoprene styrene copolymer, polybutene-1, and isobutylene isoprene copolymer.

The third component can be an ethylene based copolymer which is not, however, the same as the second component, as such would effectively make a two-component blend, rather than a three component blend as defined in the appended claims.

The polypropylene which is used as the first component of the blend provides strength to the sheet structure as well as the capability to withstand high temperature without excessive softening. The inclusion of the second and third components in the blend composition provides resilience to the blend composition.

Any of the polypropylenes may be used in the first component of the blend. Polypropylenes which are copolymers having about 2 to about 8 mole percent ethylene are preferred, as the copolymer provides some minimum level of additional resilience to the polypropylene, as compared to a homopolymer. The term "polypropylene" as used herein is intended to include homopolymers and copolymers except where specified otherwise. Whether the polypropylene is a homopolymer or copolymer, its resilience as demonstrated herein, whether having been retort processed, or not retort processed, is enhanced substantially by the incorporation of the second and third components. Without the incorporation of the second and third components, the polypropylene is at least somewhat rigid, whether it is a homopolymer or copolymer.

For example, a blend of 60% by weight of the polypropylene and 40% polyisobutylene is superior to the same polypropylene when tested in unblended composition, as measured in free fall drop tests of packages made therefrom. But these packages with polyisobutylene will fail, at a 9 foot (2.74 m) drop with the failure being by rupture through the package wall adjacent a heat seal.

By comparison, packages were made using single-layer film whose composition was 60% by weight of the same polypropylene copolymer, 20% TAFMER A and 20% polyisobutylene. The packages passed the 9 foot (2.74 m) drop tests, unlike packages made from either of the 2-component blends.

While polypropoylene homopolymer is fully satisfactory for many uses, the polypropylene copolymer is preferred, as it evidences the optimum properties of improved resistance to shock abuse in combination with the capability to withstand elevated processing temperatures. Optimum performance is generally seen, then, in blends of polypropylene copolymer with the second and third components.

While the polypropylene may be present in an amount of about 10% to about 95% by weight, a generally preferred range is about 40% to about 70%. A lower level of about 40% is generally preferred in order to maintain at least a minimal degree of the high heat temperature tolerance of the composition, and a significant moisture vapor barrier property which are provided by polypropylene. An upper level of 70% is generally desired.

While the incorporation of virtually any amount of the second component into the blend composition will provide some benefit, generally, improvements are first evident at a level of about 2% by weight. And while up to about 70% of the second component may be used, the most desired balance of properties is achieved when the second component is present in an amount of about 5% to about 35%.

Likewise, while the incorporation of virtually any amount of the third component into the blend composition will provide some benefit, generally improvements are first evident at a level of about 2% by weight. And while up to about 60% of the third component may be used, the most desired balance of properties is achieved when the third component is present in an amount of about 10% to about 40%.

The inter-relationship among the three components is a mystery, in that the use of the second and third components as defined herein may provide a blend having superior properties as compared to to the prior art. For whatever unexplained

reason, the most preferred family of compositions of the invention is that where each of the components is within its preferred range of compositions; namely about 40% to about 70% polypropylene, about 5% to about 35% of the second component, and about 10% to about 40% of the third component.

Thus it is seen that the invention provides novel material blend compositions. The invention further provides single layer films and multiple layer films and sheet structures for use in making packages. Indeed, the invention provides heat sealed packages capable of withstanding substantial amounts of physical shock type abuse, which capability is especially noticeable after the closed and sealed packages have been subjected to elevated temperatures of up to about 250 °F (121 °C).

Claims

1. A multiple layer sheet structure comprising:

(a) a first layer (12), the composition of said first layer having (i) a first component comprising a polypropylene polymer, (ii) as a second component, a polymeric composition providing elastomeric properties, said second component comprising a first moiety of ethylene and a second moiety chosen from the group consisting of propylene and butene-1, and (iii) a third component, the composition of said third component being a modifier providing elastomeric properties; and

(b) a second layer (14) adhered to said first layer, the composition of said second layer being selected from the group consisting of polymers, paper, and metal foil.

2. A sheet structure according to claim 1, wherein the second component comprises about 60% to about 80% ethylene.

3. A sheet structure according to claim 1, wherein said second component comprises about 85% to about 95% ethylene.

4. A sheet structure according to any of claims 1 to 3, wherein the composition of the third component is selected from ethylene butene-1 copolymer, ethylene propylene copolymer, polybutylene, polyisobutylene, ethylene propylene diene monomer terpolymer, styrene butadiene styrene copolymer, styrene ethylene butylene styrene copolymer, styrene isoprene styrene copolymer, polybutylene-1, and isobutylene isoprene copolymer.

5. A package made with a sheet structure according to any one of claims 1 to 4.

6. A composition of matter which is a polymeric blend, comprising:

(a) 40 to 70% e.g. 60% to 70% by weight of a first component, comprising a polypropylene polymer;

(b) a second component, comprising an ethylene-based copolymer of ethylene and a second moiety comprising either propylene or butene-1; and

(c) a third component, different from the second component which provides elastomeric properties.

7. A composition according to claim 6, wherein said second component is present in an amount of 5% to 35% by weight of the overall composition of said blend, and optionally the third component is present in an amount of 10% to 40% by weight of the overall composition of said blend.

8. A composition according to claim 6 or claim 7, wherein the second component consists of (1) a copolymer of 85 to 95 mole % ethylene and 15 to 5 mole % butene-1 or (2) a copolymer of 60 to 80 mole % ethylene and 40 to 20 mole % propylene.

9. A composition according to claim 6, wherein the second component is present in an amount of 5% to 35% by weight of the overall composition of the blend, the third component is present in an amount of 10% to 40% by weight of the overall composition of the blend, and wherein the composition of said third component is selected from styrene butadiene styrene block copolymer, styrene-ethylene/butylene-styrene block copolymer, styrene isoprene styrene block copolymer and polyisobutylene.

10. A composition according to claim 6, wherein the second component is present in an amount of 5% to 35% by weight of the overall composition of said blend, and the second component comprises a copolymer of 85 to 95 mole % ethylene and 15 to 5 mole % butene-1.

11. A composition or film according to any of claims 6 to 10, wherein the composition of the third component is selected from ethylene based copolymers, polybutylene, polyisobutylene, ethylene propylene diene-monomer terpolymer, styrene butadiene styrene block copolymer, styrene-ethylene/butylene-styrene block copolymer, styrene isoprene styrene block copolymer, polybutene-1, and isobutylene isoprene copolymer.

FIG.1

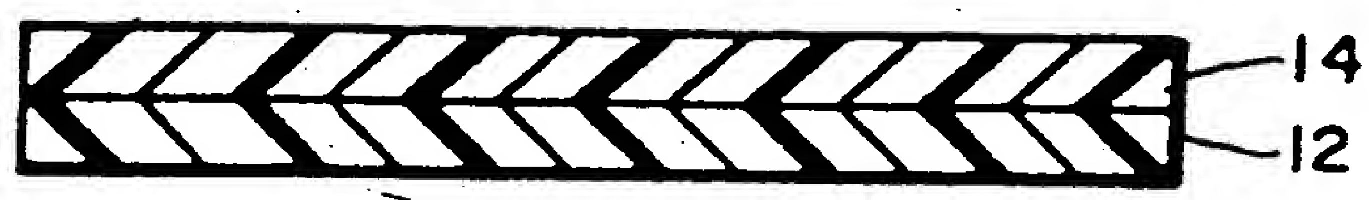


FIG.2

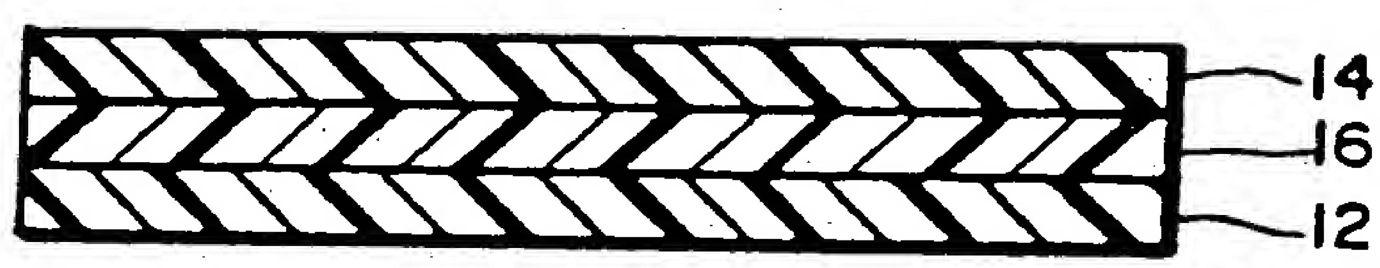


FIG.3

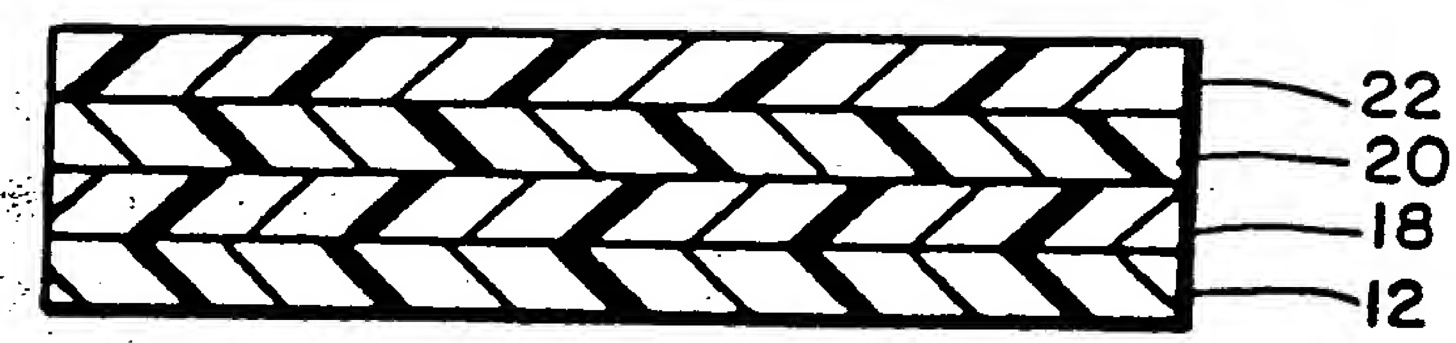


FIG.4

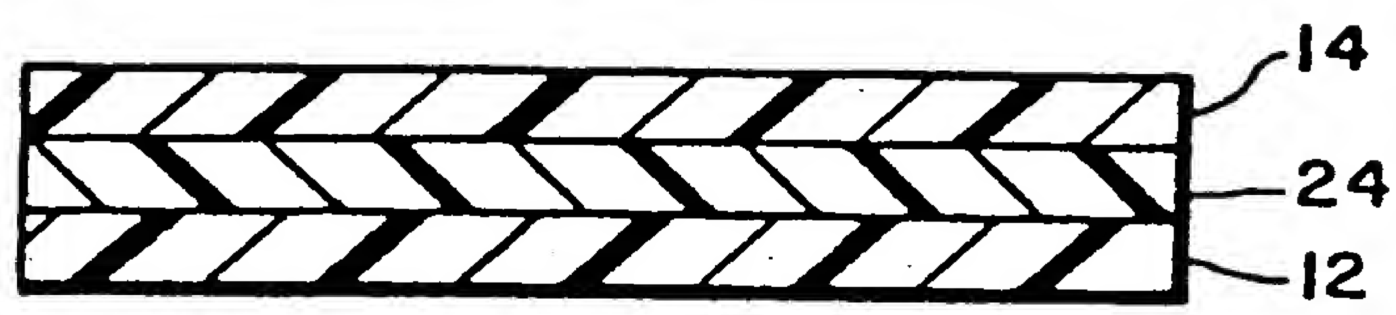
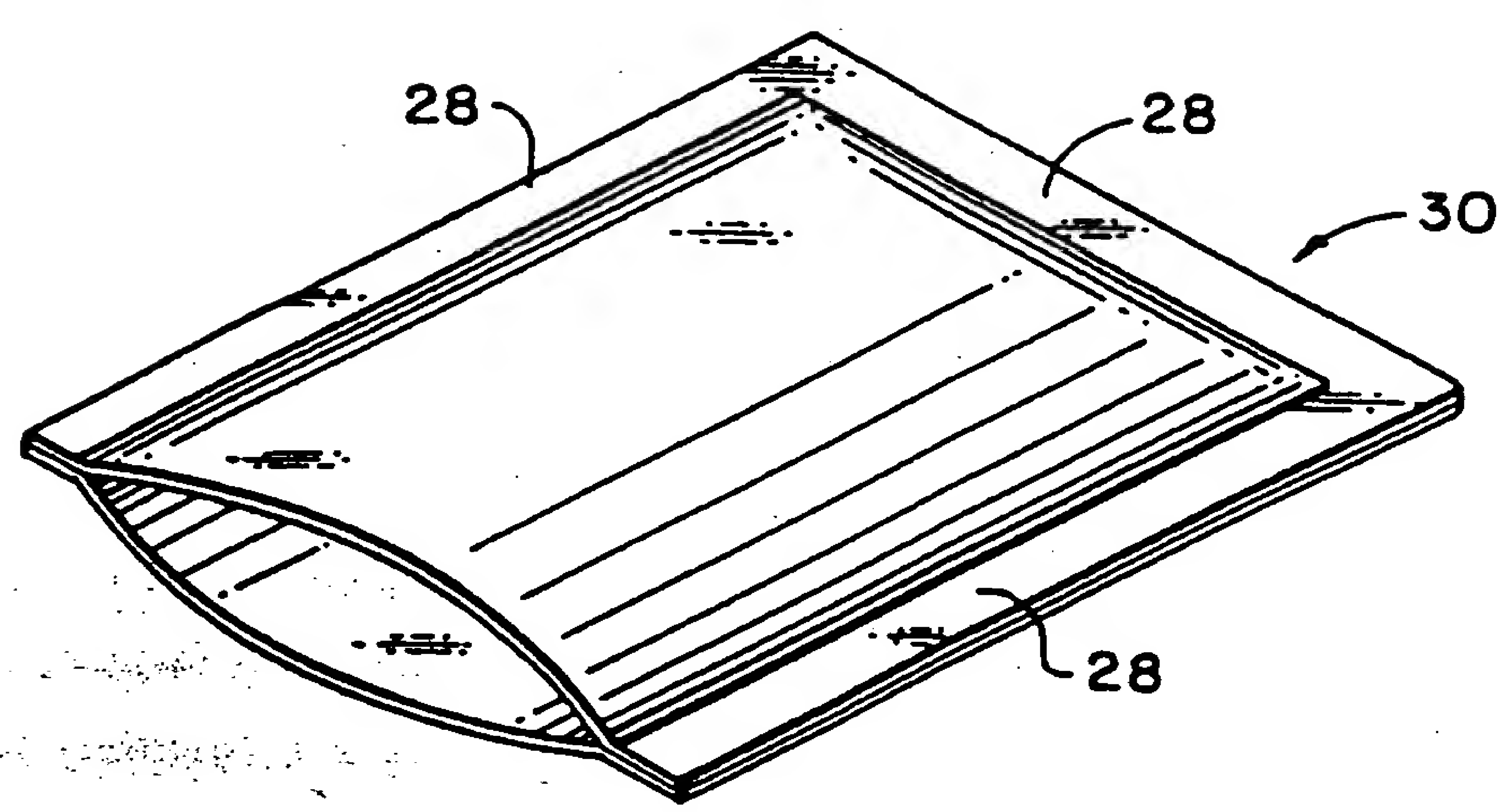


FIG.5



European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 90 10 4603

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
X	FR-A-2 415 056 (UNION CARBIDE CORP.) * Page 9, lines 1-24; claims 1,10 *	1,4,5,6	B 32 B 27/32 B 32 B 27/06 C 08 L 23/10 C 08 L 23/16 C 08 L 25/08
A		7-11	
X	GB-A-2 055 688 (TOYO BOSEKI K.K.) * Page 4, line 42 - page 5, line 8; claim 1 *	1,5,6,7,11	
A		4,8,10	

TECHNICAL FIELDS SEARCHED (Int. Cl.4)

B 32 B
C 08 L 23

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	15-04-1990	IBARROLA TORRES O.M.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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